FFLO State and Peak Effect Dynamics in CeCoIn$_5$: Magnetization Studies.

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Magnetization measurements were performed on CeCoIn$_5$ at temperature down to 30 mK with the magnetic field applied in three different orientations: parallel, near parallel ($\sim 10^0$ rotated) and perpendicular to the ab-plane. For these three orientations we have observed crossover features in the torque/magnetization traces at fields just below H_{c2}, giving further evidence for the formation of a high field Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) superconducting state in CeCoIn$_5$ for $H \parallel ab$-plane and newly indicates that the FFLO state persists for out-of-plane field orientations. Furthermore, for the (near) parallel to ab-plane field configurations and $T \leq 50$ mK, we have found an anomalous peak effect(APE) just below the crossover field when the magnetic field is sweeping down from normal to superconducting state. The dynamics of this peak suggests the existence of a metastable phase occurring in the vicinity of the FFLO phase and raises questions about the order (first or second) of the transition from FFLO to the superconducting state. None of the above features were found in a Ce$_{0.98}$Gd$_{0.02}$CoIn$_5$ crystal.

The heavy-fermion (HF) CeCoIn$_5$ is an unconventional ambient pressure superconductor with a critical temperature $T_c = 2.3$ K.[1, 2] More recently, CeCoIn$_5$ has attracted much interest owing to several unusual properties which have never been displayed by any other superconductor. For instance, a number of studies[3, 4, 5] has established that the transition at the upper critical field H_{c2} changes from second to first-order below a temperature T_0 depending on the field orientation, in contrast to the behavior of conventional type-II superconductors. This fact is considered as a strong evidence that CeCoIn$_5$ is a Pauli limited superconductor. In addition, there is growing experimental evidence that CeCoIn$_5$ is the first superconductor to exhibit the inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) superconducting (SC) state.

In the mid-1960s, theoretical studies by Fulde and Ferrell[6] and Larkin and Ovchinnikov[7] predicted a spatially inhomogeneous state to occur within the mixed state of a clean superconductor in the vicinity of the upper critical field H_{c2}. The FFLO state appears when Pauli pair-breaking overcomes the orbital effect. In the last several years, a number of type-II superconductors, including HF and organic compounds, have been claimed as likely candidates for the observation of the FFLO state. However, no experimental evidence has been accepted as an unambiguous proof for the FFLO state. With the discovery of CeCoIn$_5$, this issue has gained renewed interest. Indeed, growing experimental observations indicate the formation of the FFLO state in the high field SC phase of CeCoIn$_5$. For instance, heat capacity measurements with **H** parallel to the ab-plane have identified a second-order phase transition at $H_{FFLO}(T)$ within the SC state at low temperatures and in the vicin-

ity of H_{c2}[4, 5]. Subsequent studies, such as thermal conductivity[8], ultrasound velocity[9] and nuclear magnetic resonance[10] have given further evidence for the FFLO scenario. On the other hand, results from magnetization experiments are contradictory[3, 4, 11] and some these works have claimed absence of any anomaly associated with the FFLO state.[3, 4]

Another open issue concerns the occurrence or not of the FFLO phase for other field orientations, in particular for **H** $\parallel$ **c**. In fact, the number of studies for this orientation is considerably less than for **H** $\perp$ **c**. With the exception of the work by Bianchi et al.[5] and a more recent NMR study by Kumagai et al.[12] for whose a possible FFLO phase was identified, no evidence for the formation of such state for **H** $\parallel$ **c** has been given by other studies[3, 4, 11].

Here, we present the results of magnetization measurements performed on single crystals of CeCoIn$_5$ and Ce$_{0.98}$Gd$_{0.02}$CoIn$_5$ at temperatures down to 30 mK. The crystals were grown by In self-flux and their phase purity and SC transition were checked, respectively, by x-rays and zero-field heat capacity experiments.

The measurement technique used in this work is different from refs [3, 11] revealing new features of the magnetization of CeCoIn$_5$. Our experiments were carried out using a a diaphragm force magnetometer[13, 14] inside a plastic diluted refrigerator operating in a 20 T SC magnet(see details in ref. [13, 14]). The magnetic force on the sample was produced by a field gradient (630 Oe/cm $\leq dH_z/dz \leq$ 1.8 kOe/cm) superimposed on the main magnetic field and the sample magnetic response was detected by a capacitance technique. The contribution of the magnetic response caused by the torque was determined by repeating the measurement

with no current in the gradient coil. From one set of runs (with and without field gradient), we were able to extract the component of the magnetization parallel to the magnetic field (M_z) by a simple subtraction of the contribution of the torque from the total response for $dH_z/dz \neq 0$. This is not clearly the case by cantilever measurements [11]. Our experimental method is similar to ref [3] but the force magnetometers are different. In the present work, the movable capacitor plate of the magnetometer is a diaphragm which gives a stronger response to the torque than the apparatus used by Tayama *et al.*[15]. The data were taken for increasing and decreasing magnetic field ($|dH/dt| \approx 35$ Oe/s) after zero-field cooling the sample from well above T_c.

Figure 1 shows the capacitance response of CeCoIn$_5$ for increasing and decreasing magnetic field at $T = 30$ mK for $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \perp \mathbf{c}$ measured with and without field gradient. When $\mathbf{H} \parallel \mathbf{c}$ (FIG.1 (a)), the response for $dH_z/dz \neq 0$ is due only to the magnetization ($\mathbf{M}$) parallel to the field direction ($\mathbf{M} = M_z$). (The capacitance response is constant as a function of the field for $dH_z/dz = 0$.) The traces show a sharp jump at $H_{c2}^{\parallel}$ due to the first-order superconducting-normal state transition (FOSNT). At lower fields we observed an hysteretic peak effect near 25 kOe which follows the same trend reported in ref[3].

For $\mathbf{H} \perp \mathbf{c}$ (FIG.1 (b)), $\mathbf{M}$ is clearly not aligned to the field direction. The response with $dH_z/dz \neq 0$ shows two contributions: one caused by the torque (measured for $dH_z/dz = 0$) and the other due to M_z (displayed in FIG. 2 (b)). For both up-sweep traces (with and without field gradient), the FOSNT manifests itself by a sharp jump at $H_{c2}^{\perp}$. The jump at $H_{c2}^{\perp}$ in the down-sweep traces is smaller. The data show also the existence of a broad peak (that we will call anomalous peak effect (APE)) at H_{APE} observed only for decreasing field. This broad peak is rapidly suppressed by thermal effects and it is absent for $T > 50$ mK. The inset presents data for the Ce$_{0.98}$Gd$_{0.02}$CoIn$_5$ crystal that we discuss later.

Figure 2 shows the high field part of the magnetization traces obtained at $T = 30$ mK for CeCoIn$_5$. The data presented here are the results of the average of 4 and 7 magnetization runs obtained for $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \perp \mathbf{c}$ respectively. The discussed features of the averaged trace are also observed for each individual magnetization trace. For both orientations, the sharp jump due to the FOSNT is clearly observed for both up and down-sweep traces. From our data, we obtained $H_{c2}^{\parallel} = 49.2$ kOe and $H_{c2}^{\perp} = 117.7$ kOe with the hysteresis width of $\Delta H_{c2} \approx 0.4$ kOe. Our values are in very good agreement with reported data in the literature. In addition, for both directions, we have observed a change in the monotonic variation of the magnetization near H_{c2}. This anomaly which was not yet reported in the literature manifests itself in the derivative (dM_z/dH) trace by a step. The observed sin-
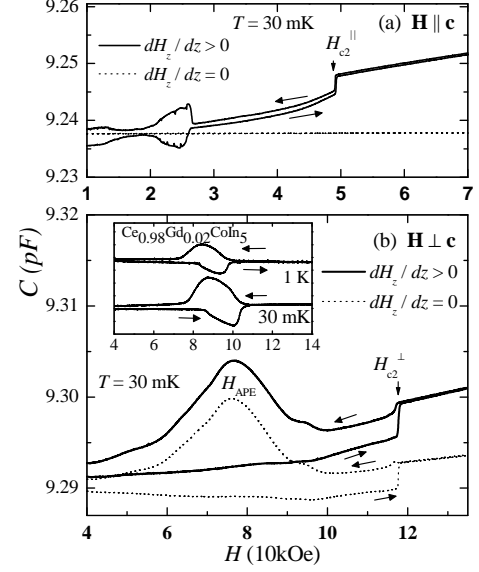

FIG. 1: Capacitance response $C(H)$ loops for CeCoIn$_5$ at 30 mK taken with and without field gradient for $\mathbf{H} \parallel \mathbf{c}$ (Fig.1 (a)) and $\mathbf{H} \perp \mathbf{c}$ (Fig.1 (b)). The inset shows similar set of data obtained at 30 mK and 1 K for Ce$_{0.98}$Gd$_{0.02}$CoIn$_5$.

gularity takes place at a field H_{FFLO} as indicated by the vertical arrows in the figure. For $\mathbf{H} \parallel \mathbf{c}$, H_{FFLO} is found to be nearly independent on T. On the other hand, for $\mathbf{H} \parallel \mathbf{c}$ the H_{FFLO} is clearly shifted to higher fields for increasing temperature.
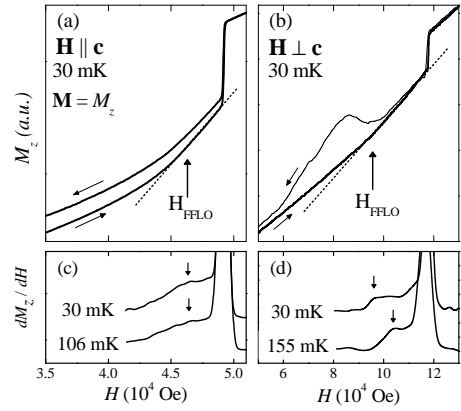

FIG. 2: High-field magnetization of CeCoIn$_5$ for $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \perp \mathbf{c}$ at $T = 30$ mK. The dotted lines emphasize the change in the monotonic variation of $M_z(H)$ near H_{c2}. For $\mathbf{H} \perp \mathbf{c}$, the APE is seen in the down-sweep trace. The respective dM_z/dH traces are shown in the Fig. 2 (c) and (d). for two temperatures. The vertical arrows indicate H_{FFLO} in both magnetization and derivative traces. The other arrows indicate the direction of the field sweep.

To further explore the existence of the FFLO phase and the nature of the broad peak at H_{APE} we have performed additional experiments. Firstly, we repeat the measurements shown in Fig.1 for the magnetic field slightly ro-

tated from the basal plane of the crystal. Figure 3 shows in the inset (a) the results for $dH_z/dz = 0$ up/down sweep traces at $T = 30$ mK and $T = 44$ mK. For $T = 30$ mK, the FOSNT occurs at $H_{c2} = 109.6$ kOe which is consistent with a misalignment of roughly $10°$ with respect to the ab-plane [11]. The H_{FFLO}-anomaly (see inset (b) in FIG. 3) was also observed in the $M_z(H)$ traces at slightly lower field than that for $\mathbf{H} \perp \mathbf{c}$ and the position of the broad peak H_{APE} is shifted to lower field as well. Secondly, we performed time relaxation measurements at selected values of the field in the region of the broad peak observed for $\mathbf{H} \parallel \mathbf{ab}$-plane and for $\theta \sim 10°$. Before each measurement, the field was sweeping up to 140 kOe, subsequently sweeping down to the target value of the field and then the signal was measured as a function of time (see inset (a) in Fig.3). The sweep rate was identical in all these measurements. The main panel of Fig.3 shows the time dependence of $C(t) \propto \tau(t) \propto M(t)$ for two fields in the region of the peak for the $\theta \sim 10°$ orientation. We concentrate on this particular orientation where the broad peak H_{APE} is even more pronounced than for $\mathbf{H} \perp \mathbf{c}$ (see Figs. 1 and 3) and the FOSNT is clearly observed for both up and down-sweep traces. But similar time-dependence was also found in the peak region for $\mathbf{H} \perp \mathbf{c}$. From the results displayed in Fig.3, we conclude that the broad peak at H_{APE} in down-sweep traces for the $\mathbf{H} \parallel \mathbf{ab}$-plane and $\theta \sim 10°$ geometries is related to a metastable state.

By mapping the T-dependence of H_{c2}, H_{FFLO} and H_{APE}, we have constructed the high-field low-temperature phase diagram for CeCoIn$_5$ displayed in FIG.4. Results from heat capacity[5] and NMR[10, 12] experiments are also shown for completeness. The evolution of the H_{FFLO} line determined from our magnetic measurements is in good agreement with the line identified by distinct techniques. Furthermore, although it takes place only at $T < 50$ mK, the H_{APE} line has the same behavior as the FFLO line.

In what follows we speculate on the origin of the APE found below the FFLO line for H (near)parallel to the ab-planes. In general, the magnetic relaxation (time dependence of the irreversible magnetization) in type-II superconductors originates from the vortex motion driven by the gradient in the vortex density ∇n_v, in presence of the vortex pinning (so-called vortex creep regime). For our studies in the (nearly) parallel to the ab-planes field configurations the torque is proportional to the c-axis component of the magnetization $M_c(t)$. Then, in the vortex creep regime, the time dependent signal $C(t) \propto \tau(t) \propto M_c(t)$. Because CeCoIn$_5$ is anisotropic (layered) superconductor, the appearance of vortex kinks along the c-axis and their interaction with vortex systems parallel to ab-planes should also be taken into account; this would provide the information on pinning of both in- and out-of-plane vortex systems[23]. In principle, our observation of the logarithmic time relaxation (see Eq. 1 below) of
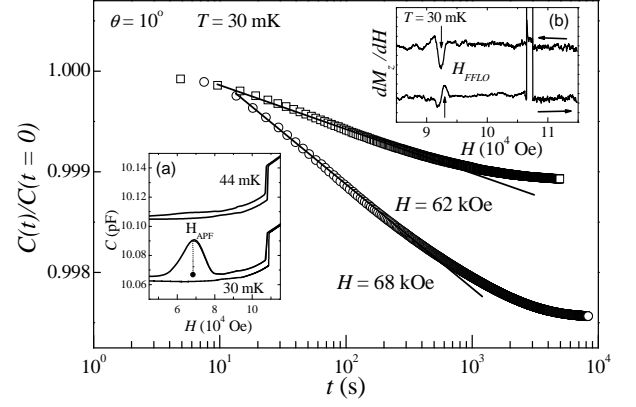

FIG. 3: Results of $C(t)$ for CeCoIn$_5$ obtained at $T = 30$ mK for two values of magnetic field applied $\sim 10°$ from the ab-planes. (a) $C(H)$ at two temperatures. The dashed line at the field H_{APE} denotes the time decay $C(t)$. (b) Up and down traces of dM_z/dH as a function of field at 30 mK. The arrows indicate H_{FFLO} for the two sweep directions.
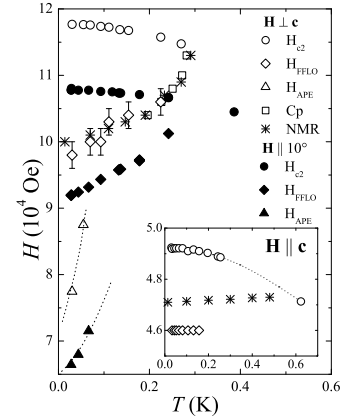

FIG. 4: $H - T$ phase diagram of CeCoIn$_5$ for $\mathbf{H} \perp \mathbf{c}$, $\mathbf{H} \parallel 10°$ and $\mathbf{H} \parallel \mathbf{c}$. The graph includes results from heat capacity[5] and NMR[10, 12]. Dotted lines are guides to the eyes.

the measured signal $C(t) \sim M(t)$ (FIG. 3) in the peak region 55 $kOe \leq H \leq 80$ kOe for the $\theta \sim 10°$ orientation, agrees with the above scenario, and suggests that the vortex pinning efficiency is enhanced in this field interval. However, in contrast to the more conventional peak effect behavior observed for $\mathbf{H} \parallel \mathbf{c}$ (see Fig. 1(a) and Refs.[3]), the APE measured for $\mathbf{H} \perp \mathbf{c}$ and $\mathbf{H} \parallel 10°$ (see Fig. 1(a) and Fig. 3), takes place only under field decreasing at very low temperatures $T \leq 50$ mK. Besides, there is an order of magnitude difference in the effective activation energy values $U_e(H, T)$ as obtained from the Kim-Anderson equation [16]:

$$M(t) = M_0(1 - \frac{k_B T}{U_e})\ln(\frac{t}{t_0}) \qquad (1)$$

Taking characteristic value $\ln(t/t_0) = 15$ [16], we find for the two field geometries in the peak effect regions $U_e(H \perp c, H_{peak} = 6.84$ $T) \sim 10^3$ K and $U_e(H \parallel c, H_{peak} = 2.46$ $T) \sim 10^4$ K. Taking into account

the relatively small anisotropy $\gamma = (m_c/m_{ab})^{1/2} \sim 2$ ([17] and refs. therein) as well as nearly the same reduced fields $(H_{peak}/H_{c2})_{||c} = 0.5$ and $(H_{peak}/H_{c2})_{\perp\ c} = 0.58$, our results suggest essentially different mechanisms responsible for the peak effects measured in $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \perp \mathbf{c}$ field configurations.

In attempts to shed light on origin of the APE, we note that irreversible under increasing/decreasing field state, characterized by a glassy-like time relaxation, has been also reported for others spin-paramagnetically limited superconductors such as Al [18, 19, 20, 21] and Be [22] films. Interestingly, the low temperature/high magnetic field portion of the $H - T$ plane (see Fig. 4), resembles very much the metastable $H - T$ phases identified in Al and Be films, where SC and normal (N) states coexist.[18, 19, 20, 21, 22]

All these may indicate that the pronounced magnetization time relaxation occurring in CeCoIn$_5$ for $\mathbf{H}$ nearly parallel/parallel to the ab-planes may be governed by dynamics of coexisting SC and N domains, or possibly BCS-like superconducting (vortex state (VS) in our case) and FFLO states. Such possibility has been considered in the context of superfluidity of atomic fermion gases.[24] Based on the experimental data of Fig. 1(b), the coexistence of N and SC domains cannot be ruled out for $\mathbf{H} \perp \mathbf{c}$ geometry, indeed. However, the results presented in Fig. 3 for the inclined field indicate that the APE takes place well outside of the field hysteresis region associated with the FOSNT. Thus, VS-FFLO domain coexistence looks a more suitable scenario. The domain boundaries can act as additional pinning centers for vortices leading to the APE. If our interpretation is correct, the FFLO line may represent in fact a first order transition, as theoretically predicted[25], giving rise to a metastable state with VS-FFLO domain coexistence in its vicinity. Whereas specific heat measurements[5] suggest the second order VS-FFLO transition, quenched disorder can mask the first order nature of the transition[26]. In fact in the closest-to-equilibrium magnetization data which is the up-sweep trace curve for $\theta \sim 10°$ orientation (see inset (b) Fig. 3), the peak-like (rather than step-like (Fig. 2)) character of the dM_z/dH anomaly at H_{FFLO} suggest a first order FFLO-VS transition. Finally, if all the features discussed in the text are indeed associated to the FFLO state, they should disappear as sample is driven away from the clean limit. To verify this idea we have performed similar experiments for Ce$_{0.98}$Gd$_{0.02}$CoIn$_5$ single crystal (inset of Fig. 1). According to our data, the substitution of Gd suppresses the FOSNT at $H_{c2}^{\perp}$. Simultaneously, the FFLO state ($\mathbf{H} \perp \mathbf{c}$) vanishes, the peak effect becomes conventional and remains to higher-T, and magnetic relaxation is invisible for the same temperature and time window (not shown) as measured for pure CeCoIn$_5$.

In conclusion, by torque/magnetization measurements we have found crossing over anomalies at the vicinity of

H_{c2} that are in agreement with $H_{FFLO}(T)$ line determined by others[4, 5, 8, 9] for $\mathbf{H} \perp \mathbf{c}$. Besides, our data indicates that the FFLO state persists for out-of-plane field orientations and reveals an APE, possibly associated with a metastable new phase below the FFLO line. This results suggest that the transition at the FFLO line may be indeed weakly first order. We hope that the new results reported in this work will stimulate further experimental and theoretical studies in the vicinity of FFLO phase in CeCoIn$_5$.

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